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# Dedicated Monte Carlo Procedures Applied in Gamma-ray Spectrometry Used in Decommissioning of Nuclear Facilities

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Additional information is available at the end of the chapter

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## Abstract

Because the experimental efficiency calibration is limited to several measurement geometries and cannot be applied directly to all measurement configurations, the efficiency transfer method for the efficiency computation was applied using ETNA software. An approach using efficiencies measured with point sources combined with theoretical procedures was applied for obtaining the peak efficiency  $\varepsilon(E)$  for disk sources measured with a NaI(Tl) detector. The transfer method was applied for the computation of the efficiency of an HPGe detector using as a reference a point source placed at 10 cm height from the face of the detector. The method was applied both for point sources and volume sources with varied compositions and densities. To correct the experimental values of the efficiencies, coincidence summing effects were evaluated using GESPECOR Monte Carlo code. The study of the response function characterization of the ISOCART and Segmented Gamma Scanner WS1100 gamma-ray spectrometry systems was related. GEANT 3.21 Monte Carlo code was used to simulate the spectra expected to be obtained for the photon energy range from 50 to 2000 keV. A big volume represented by a 220l cylindrical source was considered to be measured with the two systems. The full energy peak efficiency and the total efficiency were evaluated.

**Keywords:** gamma-ray spectrometry, Monte Carlo simulations, NaI(Tl) and HPGe detectors, efficiency transfer, radioactive waste assay

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## 1. Introduction

The decommissioning of nuclear facilities is a topic of great interest to many Members States of IAEA because a large number of facilities have to be retired from service. The term “decom-

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decommissioning" is defined in the TRS 267 [1] as actions taken at the end of a facility useful life to retire the facility from service in a manner that provides adequate protection for the health and safety of the workers, public and environment. It is a complex process because it involves many operations such as detailed survey, decontamination and dismantling of power plant equipment and facilities, buildings and structure demolition, and managing the resulting waste and other materials that need to be taken into consideration, due to their effects on health and safety of the operating personnel, public and the environment. The decommissioning activities have expanded in the last years all over the world because many nuclear installations have been exhausting their lifetime. Careful planning and management are essential to ensure that decommissioning is fulfilled in a safe and cost-effective manner. A right evaluation of the radioactivity is very important affecting directly the starting point of the decommissioning process. This can be the reason for unwanted delays between stages.

The characterization of the radioactive inventory in decommissioning wastes is described in TRS 267 [1] as a front-end task required to define the operational decommissioning plan and estimate costs and radiological risks associated with the plan. Once the decommissioning process is under way, regulatory, safety and waste disposal considerations require that the radioactive waste should be monitored and characterized. The objective of this characterization is to ensure that the waste will be handled and disposed of in a safe and economic manner. The methods and equipment used to characterize the radioactive waste resulted from decommissioning vary considerably, depending upon the type and complexity of the facility and the radionuclide mix from the plant.

The work detailed in this chapter explores the specific gamma-ray spectrometry phenomena in different work conditions, relating the analysis, development and implementation in the radioactive waste management of specific investigation methods for gamma-ray spectrometry measurements that will produce reasonable measurement uncertainties [2] with lower cost and relatively short duration of data acquisition. The applicability and functionality of gamma-ray spectrometry methods to radiological characterization and free release of radioactive waste materials are presented, using experimental methods that are mostly combined with theoretical and simulation procedures using Monte Carlo computer codes.

## **2. Efficiency transfer in gamma-ray spectrometry**

Because more and more nuclear installations reach the end of their life, the dismantling and decommissioning processes of them became a key topic in the nuclear industry. The radiological characterization of the systems, structures, equipment, components and the environment represents a basic phase in the decommissioning process because allow the definition of the decommissioning strategy. This task is very important because it provides the basis for the correct classification of various types of waste, which in turn affects the decommissioning solution and the associated costs. The measurement method should be reliable and efficient. In addition, it should be flexible, able to provide proper results for the diversity of samples assessed with different compositions and densities, different shapes and possibly non-uniform

activity distribution. The appropriate efficiency calibration in the deprived conditions is a challenging task.

The experimental calibration of germanium detectors used in gamma-ray spectrometry [3] is difficult to achieve particularly for the geometry measurement that cannot be estimated as a point source. Therefore, when different samples with varies composition and densities are measured in various geometries, a large number of standards need to be measured to carry out the detection efficiency according to a specific sample matrix and geometry. The situation is more difficult in the case when the samples are measured with high efficiency detectors and the sample is placed close to detector, because in this case most radionuclides will give rise to important coincidence summing effects [4]. Consequently, the detection efficiency for a given energy depends not only on energy and experimental setup, but also on the radionuclide. The knowledge of the detection efficiency, which varies strongly with the source to detector distance, due to the geometry and absorption factors, is essential for operating these systems. Therefore, a comprehensive experimental calibration would require the measurement of a big number of standards, one for each geometry and matrix of interest, containing certified activities for each radionuclide that is present in the real samples. A better solution for determining the detection efficiency is the application of specific methods of calculation. In the gamma-ray spectrometry field, except the simulations performed for the calculation of the detection efficiencies, Monte Carlo simulation codes can also be used to evaluate the transfer factors [5]. Based on its relative sensitivity to the uncertainties of the detector parameters and of the calculation model, the method of efficiency transfer [6, 7], based on Monte Carlo simulation or on semi-empirical methods [6, 8, 9], is more and more relevant to evaluate the efficiency whenever direct experimental calibration is not accessible [10].

The applicability of the ETNA (Efficiency Transfer for Nuclide Activity) software to compute the efficiency transfer factors for various counting geometries used in routine laboratory measurements was examined. Thus, ETNA results were compared with experimental results (corrected for coincidence summing effects) [10, 11]. The detection efficiencies were calculated for NaI(Tl) and HPGe detectors. An approach using the experimental efficiency measured with point sources combined with theoretical procedures was applied for obtaining the peak efficiency  $\varepsilon(E)$  for disk sources measured with NaI(Tl) detector. Using the detection efficiencies for a reference point source geometry located at 10 cm distance from the high purity germanium (HPGe) detector, the applicability of the efficiency transfer method was checked once more.

## 2.1. The efficiency calibration of the detectors using experimental measurements

For the efficiency evaluation of the disk sources, the measurements were made with an Ortec gamma-ray spectrometry system consisting of a ScintiPack Photomultiplier Base with Preamplifier and High Voltage Supply type 296 and a DigiDART Digital Portable Multichannel Analyzer and lead collimator. The NaI(Tl) detector specifications are as follows: the diameter of the end cap of the detector is 3 × 3 inches, the crystal diameter is 8 cm, and the energy resolution is 70.62 keV at 1332 keV ( $^{60}\text{Co}$ ). The recommended operating bias is +1000 V.

The second Ortec gamma-ray spectrometry system used in this study for the examination of the applicability of the efficiency transfer method consisted of a high purity germanium detector, model GMX50P4, transplantable in Pop Top technology, with dimensions: 6.46 cm diameter, 7.5 cm length, 0.05 cm beryllium absorber layer and a Digital Portable Multichannel Analyzer type DigiDART. The main performance specifications of the HPGe warranted by the producer are presented in **Table 1**.

Resolution (FWHM) at 1.33 MeV, <sup>60</sup> Co	2.2 keV
Peak-to-Compton ratio, <sup>60</sup> Co	58:1
Relative efficiency at 1.33 MeV, <sup>60</sup> Co	50%
Peak shape (FWTM/FWHM), <sup>60</sup> Co	2.0
Resolution (FWHM) at 5.9 keV, <sup>55</sup> Fe	800 eV
Recommended operating bias, negative	3300 V

**Table 1.** HPGe performance specifications.

In the first step, certified standard point sources were used to evaluate the detector experimental efficiencies as a function of gamma-ray energies [3], for the NaI(Tl) detector.

The sources were measured in horizontal plane, at radial distances  $r = 0, 1, 2, 3$  and  $4$  cm from the detector axis and at  $0.8$  cm from the face of the NaI(Tl) detector. A lead collimator was used in the measurements. Five sets of data were obtained for all the important gamma line involved in the study. From the graphic representation (**Figure 1**), it can be seen that the experimental efficiencies  $\varepsilon(E)$  for the NaI(Tl) detector do not present a smooth variation with the energy  $E$ .

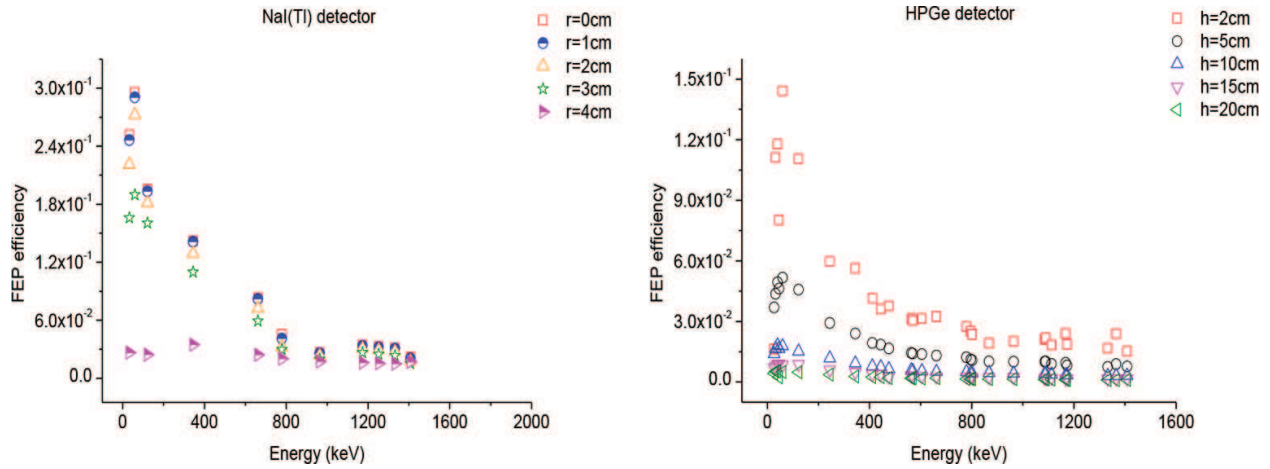
In the case of the HPGe detector, the experimental detection efficiencies were evaluated for the detector-point sources distances of  $2, 5, 10, 15$  and  $20$  cm. The counting dead time of the measurements was in general controlled to be less than  $7\%$  and consequently corrected during the counting. The amplifier time constant was fixed to  $12\ \mu\text{s}$ . Cylindrical sources (**Table 2**) were also measured at  $0, 1, 2$  cm from the face of the HPGe detector. The counting dead time for these sources was less than  $3\%$ .

Radionuclide	H (cm)	D (cm)	$\rho$ (g/cm <sup>3</sup> )	$\Lambda$ (Bq)	$u$ (%) ( $1\sigma$ )
<sup>134</sup> Cs	3.2	7.4	1.0	1916	2.5
<sup>137</sup> Cs	3.3	7.4	1.4	1190	3.5

**Table 2.** Cylindrical sources.

<sup>134</sup>Cs and <sup>137</sup>Cs radioactive sources were used to test the applicability of ETNA software for volume sources. Water and soil matrix have been chosen because they are most common in gamma-ray spectrometry laboratory.

The directly measured efficiency calibration curves obtained for the HPGe detector are represented in **Figure 1**. Was observed that the experimental efficiencies values  $\varepsilon(E)$  for the HPGe detector do not present a smooth variation with the energy,  $E$ .



**Figure 1.** The experimental values of the detection efficiencies for point sources measured with the NaI(Tl) and HPGe detectors.

The uncertainties ( $1\sigma$ ) of the experimental efficiencies values were estimated using ISO standard [12] using uncertainties of the activities from the certificates and the uncertainties of the counting results. The values were up to 11% for  $r=0, 1, 2, 3$  and 4 cm distances from the NaI(Tl) symmetry axis and up to 3% for  $h=2, 5$  and 10 cm and up to 8% for  $h=15$  and 20 cm source-to-detector distances in the case of the HPGe detector.

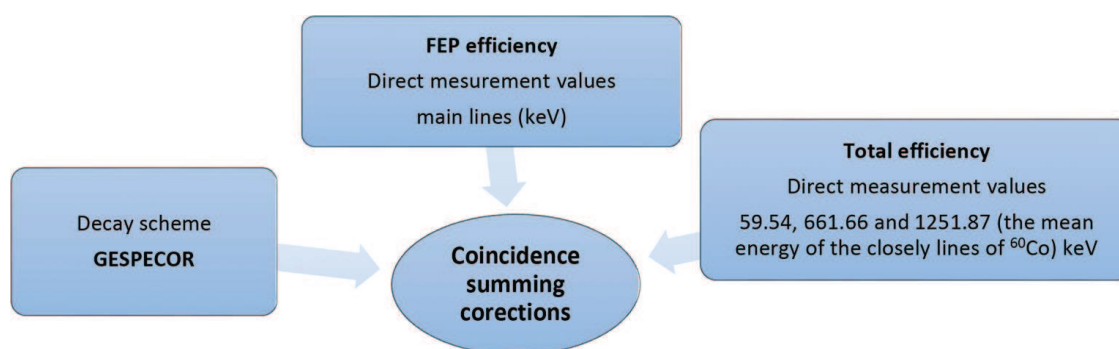
## 2.2. Coincidence summing corrections

The origin of the efficiency data deviation from smooth curves as a function of energy is due to the presence of significant coincidence summing effects in the case of  $^{152}\text{Eu}$ ,  $^{134}\text{Cs}$  and  $^{60}\text{Co}$  sources [10, 11]. To remove the effects of coincidence summing, specific corrections were evaluated and applied to experimental efficiencies for the purpose to obtain useful efficiency curve. To evaluate the coincidence summing effects, it represents a difficult task, mainly when the nuclides present complex decay schemes. To obtain the correct efficiencies values for the  $^{152}\text{Eu}$  energy lines, the peak and total efficiencies are required for the energies of supplementary photons emitted by  $^{152}\text{Eu}$  nuclide. For instance, in the case of the peak with energy  $E = 121.78$  keV, only coincidence losses are feasible. The process is produced when any photon from the 71 photons list is emitted instantaneously with the photon with  $E = 121.78$  keV and together interacts with the detector. These photons span an energy range from  $\chi$ -ray to  $E = 1647$  keV, and therefore, the total efficiency for the energy in this range is needed. For the peak with  $E = 1408.01$  keV, 21 moistures of various photons are possible and can contribute to sum peak effects. To evaluate the coincidence summing effects for the  $E = 1408.01$  keV, the peak efficiency for all these photons is required.



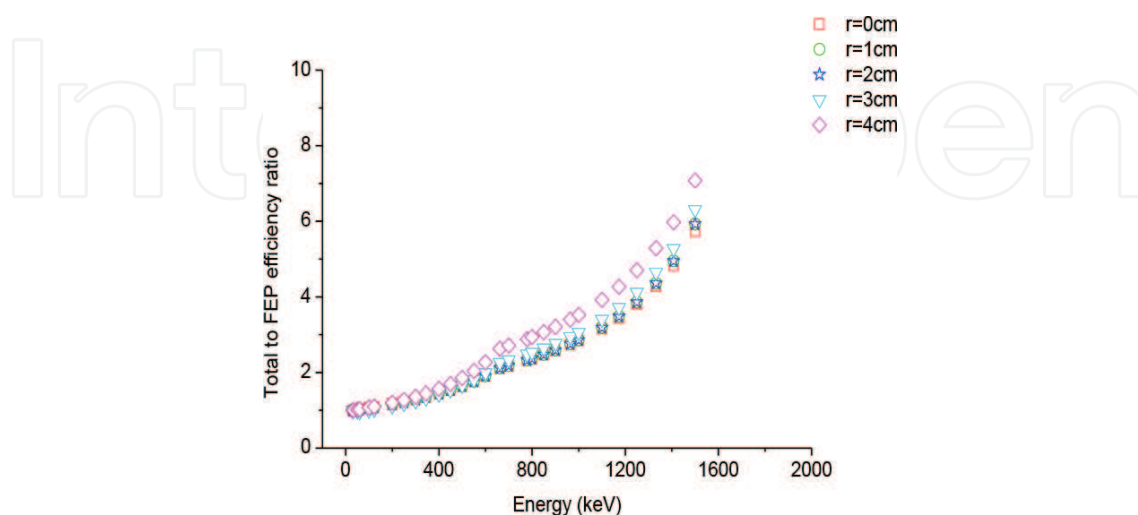
A method available to correct these effects is the Monte Carlo method. A dedicated software called GESPECOR [6] has been applied, in order to evaluate the coincidence summing corrections. This is user-friendly Monte Carlo software useful for the computation of the efficiency [3], of matrix effects [13] and of coincidence summing corrections [4] in gamma-ray spectrometry with HPGe detectors.

Because GESPECOR is dedicated to germanium detectors, the code cannot be directly applied for NaI(Tl) detector. Therefore, in the case of NaI(Tl) detector, the coincidence summing correction factors have been evaluated using an iterative procedure. Both the decay scheme data evaluated by GESPECOR and the experimental values of the peak and of the total efficiencies for the point source measurements were needed. The procedure followed in the first iteration is represented in **Figure 2**.



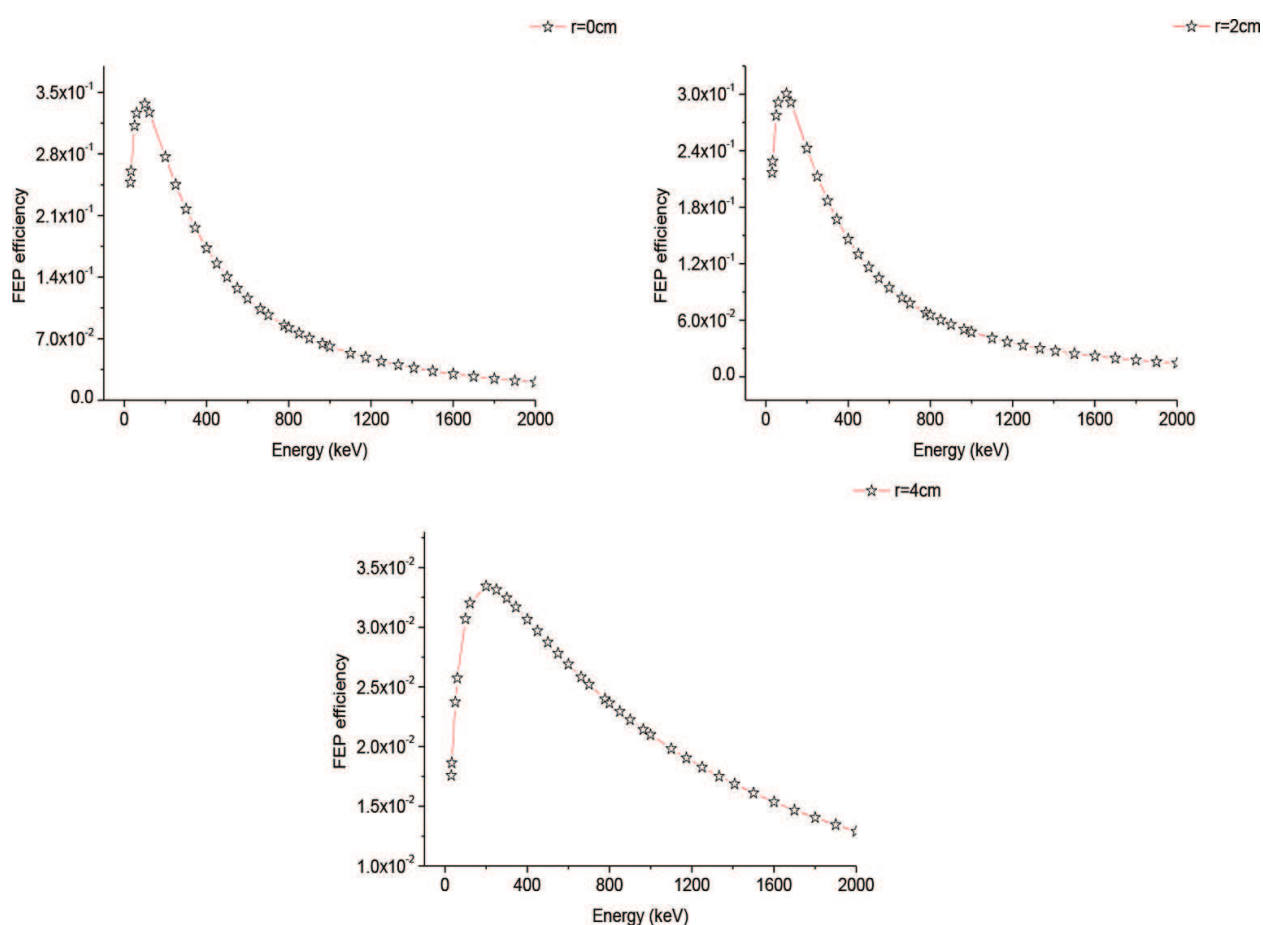
**Figure 2.** Procedure for coincidence summing correction factors evaluation

Taking into account that the ratio between the peak and total efficiency is a smooth function of energy, a first estimate of the total efficiency as a function of energy was obtained (**Figure 3**). This was possible even if only few directly measured total efficiency data were available.



**Figure 3.** The experimental ratio of total to peak efficiency versus the energy for the NaI(Tl) detector.

The obtained values of the peak and total efficiencies were used to calculate the coincidence summing correction factors in the first iteration. The correction factors were subsequently used to obtain improved values of the peak efficiencies for the NaI(Tl) detector (second iteration). The peak and total efficiencies values resulted in the second iteration obtained using the similar procedure followed in the first iteration procedure used to evaluate the coincidence summing correction factors in the second iteration. Was observed that it was not necessary to proceed in a higher order iterations. The final values obtained for the peak efficiencies of the NaI(Tl) detector were achieved from the measured values of the peak efficiencies and the coincidence summing correction factors in the second iteration. In **Figure 4** are represented the detection efficiencies for point sources measured in horizontal plane at 0.8 cm from the end face of the detector, corrected for the effects of coincidence summing for NaI(Tl) detector, only for  $r = 0, 2, 4$  cm.

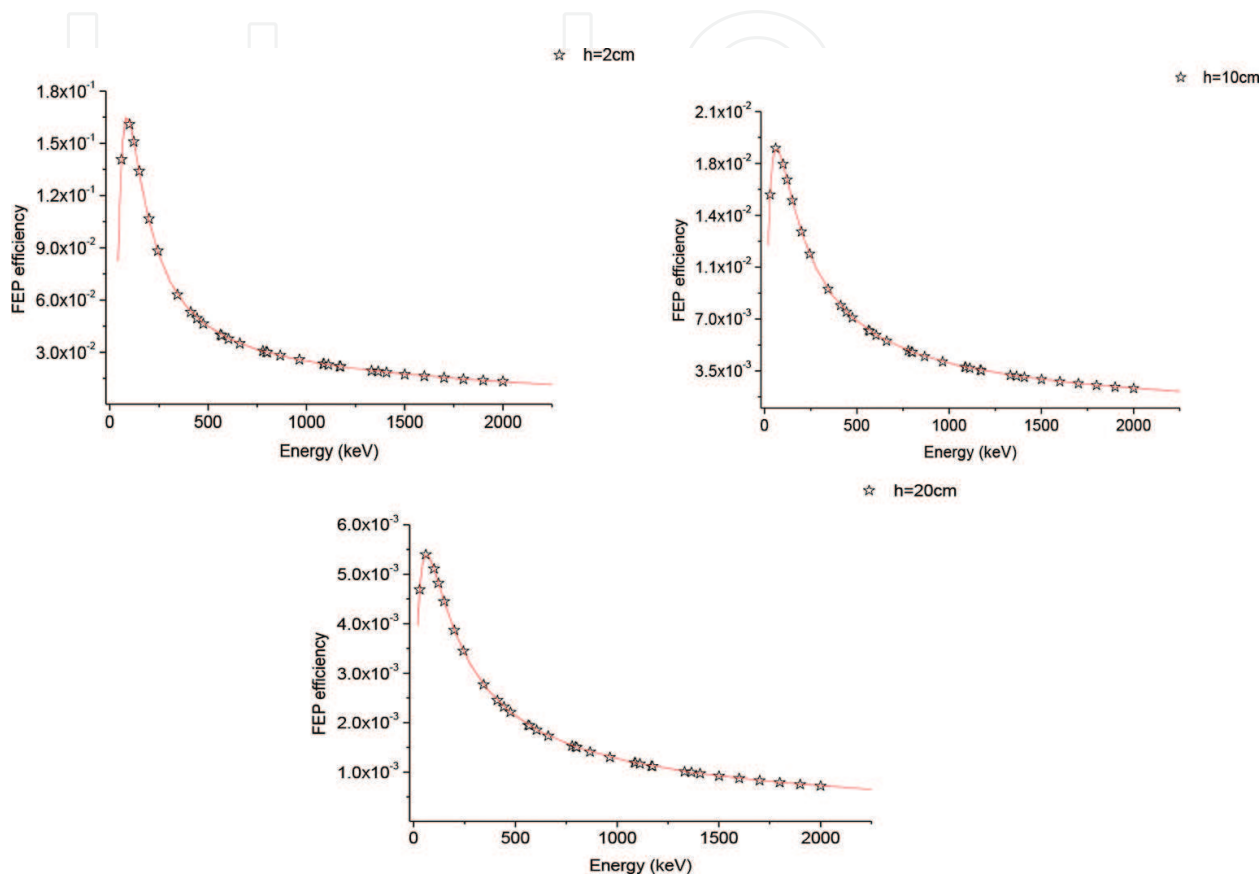


**Figure 4.** The detection efficiency corrected for the effects of coincidence summing for NaI(Tl) detector.

In the case of HPGe detector, the coincidence summing effects are presented in the case of  $^{152}\text{Eu}$ ,  $^{134}\text{Cs}$  and  $^{60}\text{Co}$  sources. Specific coincidence summing corrections were applied to the experimental efficiencies in order to obtain a generally useful efficiency curves for the HPGe detector. The values included in the HPGe detector manufacturer's data were used in computation as input detector data. The computed correction factors were subsequently used to



obtain improved values of the peak efficiencies. In the case of cylindrical sources, the corrections were necessary only for  $^{134}\text{Cs}$  gel matrix. The detection efficiency curves (corrected for the effects of coincidence summing) in function of energy for the five source-to-HPGe detector distances obtained for the peak efficiency for point sources are represented in **Figure 5** only for  $h = 2, 10, 20$  cm.



**Figure 5.** The detection efficiency corrected for the effects of coincidence summing for HPGe detector.

The efficiencies for the point sources obtained in this way for the reference measurement geometry (10 cm source-to-HPGe detector distance) could be used to evaluate the efficiency for other measurement geometries by the efficiency transfer method.

The uncertainties ( $1\sigma$ ) of the corrected efficiencies for point sources were up to 3% for NaI(Tl) detector and up to 3.5% for  $h = 2, 5$  and 10 cm and 8.5% for  $h = 15$  and 20 cm for HPGe detector. The uncertainties of the corrected efficiencies for cylindrical sources include additional uncertainties of the matrix effects; the resulting values were up to 8% ( $1\sigma$ ).

### 2.3. The efficiency transfer

The gamma-ray spectrometry method is a relative method, necessitating standard radioactive sources for the efficiency calibration. When the standard source and the sample are the same, the next relation is applied for the computation of the activity of the sample:

$$\Lambda_p(E) = \Lambda_e(E) \frac{R_p(E)}{R_e(E)} \quad (1)$$

where  $\Lambda_p(E)$  and  $R_p(E)$  are the activity and the count rate for the sample, and  $\Lambda_e(E)$  and  $R_e(E)$  are the activity and the count rate for the standard source corresponding to the peak with energy  $E$ .

In practice, it is difficult to use standard sources for all samples geometries. Accordingly, for this purpose, the efficiency transfer method can be used. Starting from the experimental efficiency for a reference point source, the efficiency for the sample can be evaluated using a mathematical model or simulation software.

Formally, the efficiency transfer method is based on the next equation [5]:

$$\varepsilon_{(calc)} = T\left(\frac{calc}{ref}\right) \varepsilon_{(ref)} \quad (2)$$

where  $T\left(\frac{calc}{ref}\right)$  is the transfer factor.

The transfer factors can be calculated with the Monte Carlo [6] method or with more simplified procedures [14, 15] using the relationship between the simulated efficiency for measurement geometry and the efficiency for the reference geometry. The benefit of this method is that the results are less affected by the uncertainties of detector parameters, which represent a more important uncertainty source in the direct simulation of the efficiencies. Undoubtedly, an improper value will be considered for the detector radius, and this will strongly affect the values of the efficiencies calculated by simulation or evaluation by analytical computation, while the transfer factor is slightly sensitive to this incorrect value, because a wrong value will simultaneously affect both the efficiency calculated for the reference geometry and that for the geometry of interest and their ratio will be practically unchanged. The sensitivity of the efficiency to details of the input data and to the computation model was clearly revealed by Vidmar in 2008 [16]. Clearly, the efficiency transfer method offers better results in the case when the measurements of interest are similar to the reference measurements.

The National Laboratory Henri Becquerel (LNHB) from Saclay, France, developed in the early 2000s a software named ETNA (Efficiency Transfer for Nuclide Activity), dedicated for the calculation of the detector efficiency under measurement conditions different from those of calibration, and for the correction of the coincidence summing effects. The application of the ETNA software makes possible to greatly increase the accuracy of the results of quantitative analysis by gamma-ray spectrometry and avoid time-consuming measurement sequences [15].

The ETNA software was applied for the evaluation of the efficiencies for various geometries based on the efficiencies values for the reference measurement geometry.

### 2.3.1. Computation of the efficiency for disk sources for the NaI(Tl) detector

ETNA software was used in this section to achieve the efficiency transfer from a point source geometry to disk sources geometries for the NaI(Tl) detector. Due to lack of calibration certificates for standard disk sources, the efficiency calibration was calculated based on point sources measured with the NaI(Tl) detector.

Assuming symmetry of the cylindrical detector, for the determination of the experimental efficiencies in view of surface contamination measurement, the next relations were used:

$$N = \int_{(S)} \varepsilon(r) \Lambda_s(r, \phi) dS \quad (3)$$

$$\approx N = \int_0^R \int_0^{2\pi} \varepsilon(r) \Lambda_s(r, \phi) r dr d\phi \quad (4)$$

$$\approx N = \int_0^R \varepsilon(r) r dr \int_0^{2\pi} \Lambda_s(r, \phi) d\phi \quad (5)$$

$$\approx N = 2\pi \int_0^R \varepsilon(r) \overline{\Lambda_s(r)} r dr \quad (6)$$

where  $N$  is the peak count rate;  $\varepsilon(r)$  is the efficiency of a point source situated at distance  $r$  from the symmetry axis of the detector;  $\Lambda_s(r, \phi)$  is the surface activity of the source in the point of  $(r, \phi)$  coordinates;  $\overline{\Lambda_s(r)} = \frac{1}{2\pi} \int_0^{2\pi} \Lambda_s(r, \phi) d\phi$ .

Considering that the surface activity is uniformly distributed, result that:

$$N = 2\pi \Lambda_s \int_0^R \varepsilon(r) r dr \quad (7)$$

$$N = \Lambda_s I(R) = \pi R^2 \varepsilon \Lambda_s \quad (8)$$

$$\text{or } \varepsilon = \frac{1}{\pi R^2} I(R) \quad (9)$$

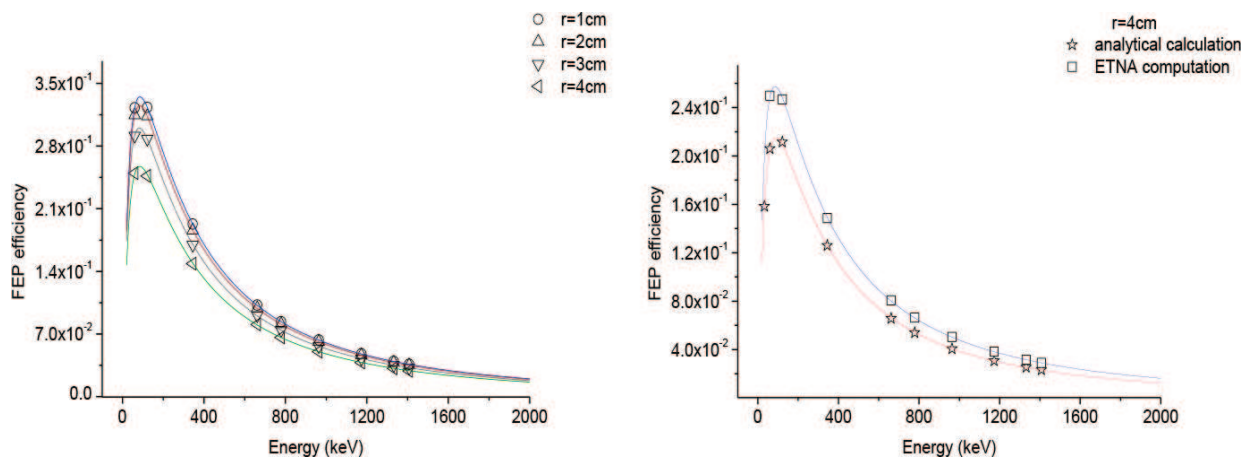
$$\text{were : } I(R) = 2\pi \int_0^R \varepsilon(r) r dr \quad (10)$$

Then, the following result is obtained:

$$\varepsilon = \frac{2}{R^2} \int_0^R \varepsilon(r) r dr \quad (11)$$

Using Eq. (11), the detection efficiency for the disk sources with radius of  $r = 1, 2, 3$  and  $4$  cm for the NaI(Tl) detector was calculated. The values of the efficiencies  $\varepsilon(r)$  were taken from the measured efficiency curves, corrected for coincidence summing effects.

Using the ETNA software, the detection efficiencies for disk sources were calculated for the same type of detector and the same measuring geometry. The reference measurement is represented by a point source located at  $r = 0$  cm (on the symmetry axis of the detector); thus, the reference efficiency is  $\varepsilon_0$ . In **Figure 6**, the efficiency calibration curves, evaluated with the ETNA software, are represented for the disk sources with radius of  $r = 1, 2, 3$  and  $4$  cm; and a comparison between the results obtained applying Eq. (11) and those evaluated using ETNA software for efficiencies detection for a disk source with  $r = 4$  cm.



**Figure 6.** The efficiency calibration curves and ETNA vs. analytical calculation for the NaI(Tl) detector.

The values obtained with the analytical procedure are in accordance with the values resulted using ETNA software. The differences can arrive from the method used or from the input data. Should be mentioned that ETNA software calculates efficiencies for geometries in which the center of the source is placed only on the detector axis; different radial distances than that cannot be included.

### 2.3.2. The ETNA computation for the HPGe detector

The transfer of the efficiency from the reference point source geometry,  $h = 10$  cm, to other point source geometries (distances from the detector end cap equal to 2, 5, 15 and 20 cm) and the computation of the efficiency for cylindrical samples with different matrices was done using ETNA software for the HPGe detector.

Using the fitted efficiency data for the reference measurement as input, the description of the reference source, the description of the source for which the efficiencies are required, the

source-detector distance, the detector-absorber distance, ETNA software was applied for the computation of the efficiency for the other measurement geometries for the HPGe detector. The experimental efficiency curves were compared with the prediction of the ETNA software. Excepting the case of the smallest distance from the source to the detector, the discrepancies between ETNA and the experimental results were generally below 3%. In the case of the measurement at 2 cm distance from the detector, the discrepancies were higher being sensitive to the detector geometrical data. This is because the detector specifications established by the manufacturer of the detector were used without any optimization. Furthermore, the uncertainty of the distance between the crystal and the end cap (the manufacturer value) has a contribution in the uncertainty of the transfer factor, because of the change in the solid angle [10].

The HPGe detector efficiency transfer method has also been used for the efficiencies evaluation for the specific cylindrical sources. For this purpose, the matrix was considered water equivalent for the matrix with  $\rho = 1.0 \text{ g/cm}^3$  and soil composition for the matrix with  $\rho = 1.4 \text{ g/cm}^3$ .

The default attenuation coefficients foreseen by ETNA code were used for the matrices involved in the study.

In the case of the soil matrix, containing  $^{137}\text{Cs}$ , the ratio between the ETNA software values and the experimental values of the efficiency was 1.038 for  $h = 0 \text{ cm}$  and 0.966 for  $h = 2 \text{ cm}$ . The higher discrepancies in the case of  $^{134}\text{Cs}$  results (gel matrix) in comparison with the results for  $^{137}\text{Cs}$  (soil matrix) can be attributed to the uncertainty of coincidence summing effects and of the matrix effects.

### 3. Simulation of gamma-ray spectra using Geant 3.21 Monte Carlo code for sources distributed in 220l volume

Application area of the radiation transport modeling through Monte Carlo method is extremely large, from the nuclear reactor design to parameters calculation of complex detection systems, from the simulation and the interpretation of various experiments to the calculation of the dose coefficients. Nowadays, this area is expanding, both by tackling new problems in modeling and by the inclusion of some details, previously neglected, of the respective phenomena [17]. What characterizes the Monte Carlo method is the remarkable fidelity with which it can describe physical phenomena, without approximations [18, 19]. Theoretically, the accuracy of results is limited only by the accuracy of the nuclear data (the effective cross section of interaction) used, and no other method is competitive with the Monte Carlo method in this regard. Accordingly, the Monte Carlo method is often used as a reference method; the simplified calculation procedures, faster, based on some approximations are tested in relation to this method, and even nuclear data can be validated by interpretation of the Monte Carlo complex experiments [18, 19].

In the Monte Carlo method, a problem of radiation transport is solved by simulating the evolution of a large number of radiations and the analysis of their fate. The evolution (the



history) of each radiation starts with its emission continues with the undisturbed propagation through the environment between the successive interactions and by changing its parameters (possibly even its disappearance) in the points where the interactions takes place. In the case of photon transport at keV–MeV energy range, complex effects are important, such as Compton photoelectric and electron–positron pairs generation effects. Following the Compton effect, the incident photon energy is transferred to the recoil electron and to the scattered photon. In the photoelectric effect, the photon is absorbed, an electron is emitted, and the atom remains in an excited state. In the pair production effect, the photon energy is consumed to produce a pair of electron–positron particles. Thus, except for the Compton effect, the history of each photon ends at the interaction point. However, in terms of energy dissipation in substance, the primary interaction of the photon is only the starting point. The resulted electrons give their energy causing ionization and excitation, emitting new photons (secondary radiation), and the atoms will relax from excited states, emitting new photons and possibly Auger electrons; the positron will annihilate producing annihilation photons that interact further. Obviously, if more details of the secondary radiation will be included, the computing time will increase, and the computation program becomes more complex with more branched. At every interaction, the evolution of the resulting products and then of the successive generations of secondary radiations should be evaluated. Which details are relevant and which are not depends on the problem to be solved for achieving an optimal compromise between modeling finesse, the required accuracy and the programming effort and the necessary computing time. For incident photons with energy up to a few MeV, characteristic X-rays, the bremsstrahlung radiation (emitted by electrons resulting from interactions) have energies much lower than the primary photon energy. Consequently, in many problems, X-rays can be considered locally absorbed.

The development of the Monte Carlo methods and the improvement of computational technologies have led to the development of several Monte Carlo simulation programs for simulating radiation transport. Simulation codes used are GEANT 3 [20], GEANT 4 [21], MCNP [22], GESPECOR [6], FLUKA [23], ETRAN [24], EGS [25–27], PENELOPE [28], etc.

In this section, the application of Monte Carlo simulation to the study and examination of the response function characterization of two gamma-ray spectrometry systems used for measuring large sources was reported. For this purpose, GEANT 3.21 code was applied for the spectra simulation expected to be obtained for 50–2000 keV energy range for volume sources measured with both systems. Although the prevalent application of Monte Carlo simulation for efficiency calibration of HPGe detectors for the measurements of small volume samples up to several dm<sup>3</sup>, extensive realistic computations by Monte Carlo methods have not been carried out until now for the measurement of big volume samples like 220l waste drums.

### 3.1. Experimental configuration

The first system used is an ISOCART from Ortec (Geom1) and has a p-type detector with a relative efficiency of 25%. The second system used is a WS1100 Segmented Gamma Scanner from Canberra (Geom2) and has a p-type detector with a relative efficiency of 44.4%. The characteristics and dimensions of the detectors are presented in **Figure 7**.



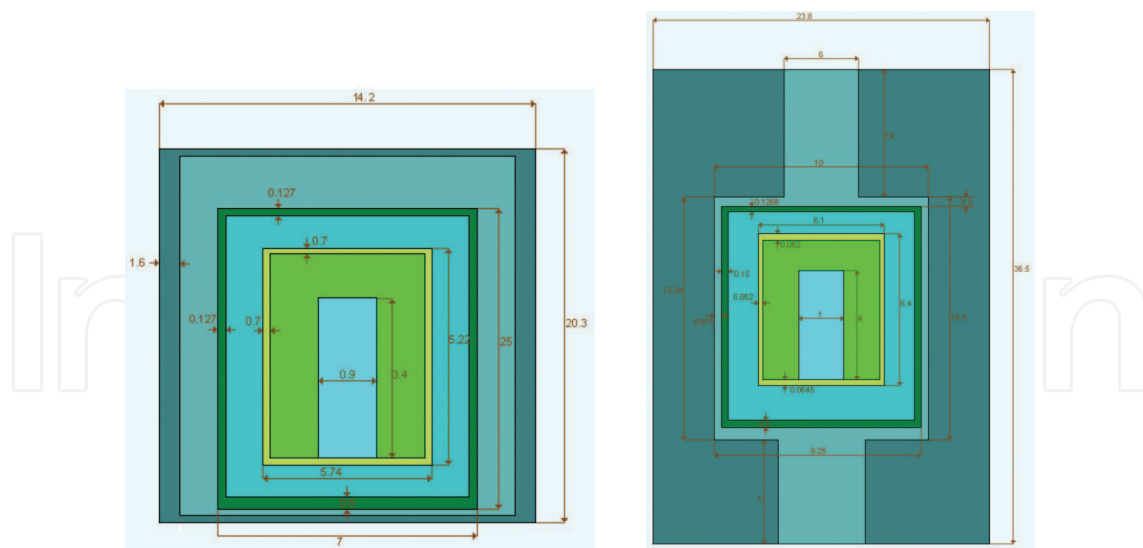


Figure 7. ISOCART (left) and Segmented Gamma Scanner WS1100 (right) gamma-ray spectrometry systems.

The volume source considered in simulation was a 2201 radioactive waste drum typically used for conditioning of radioactive waste in Romania. Several studies were reported using this kind of sample [29–32]. The source matrix was considered to be concrete with standard composition, and the axis of the detector was perpendicular on the axis of the cylinder. The distance from the center of the coordinate system associated to the detector to the center of the cylinder was 50 cm for both geometries.

3.2. Monte Carlo simulations

GEANT 3.21 [20] is a method of detector description and simulation tools, whit characteristics presented in Figure 8.

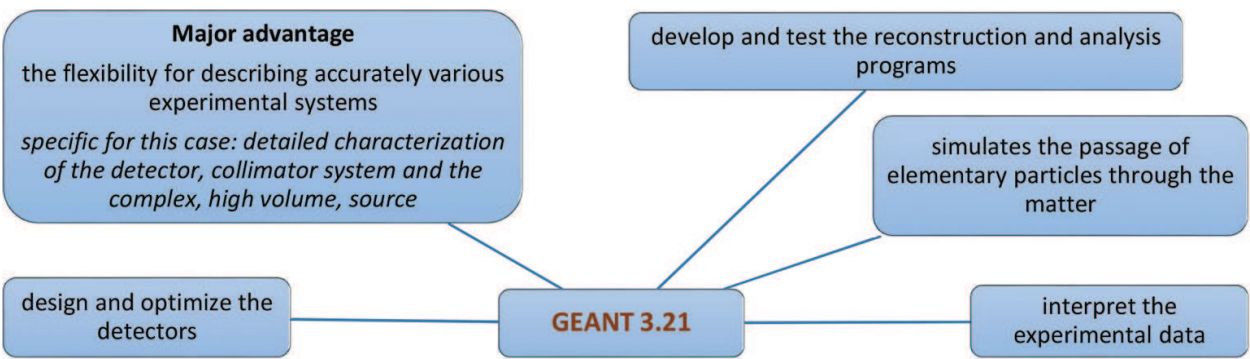
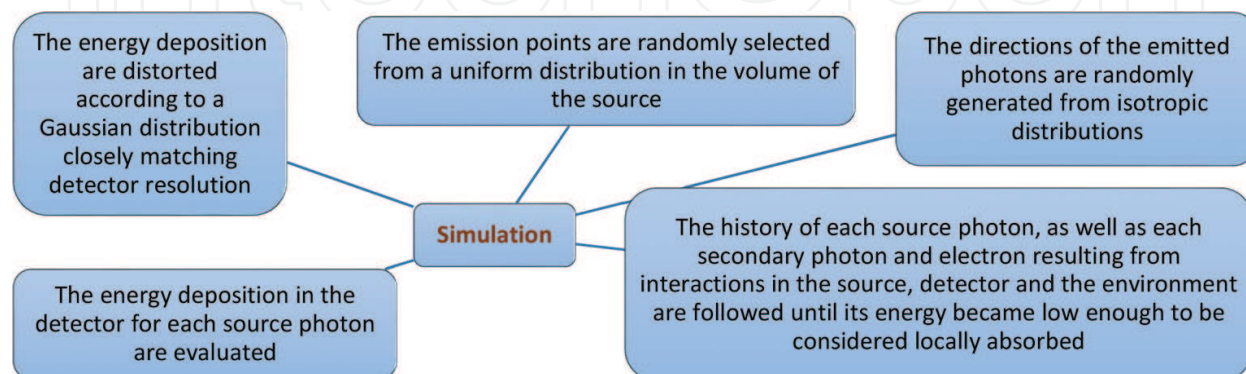


Figure 8. GEANT 3.21 characteristics.

In essence, the utilization of the Monte Carlo simulation method for the detection efficiencies evaluation for large samples such as waste drum is nearly the same as in the situation of small samples. Considering the practical aspects, it appears a large difference regarding the calculation time. In the case of big samples, in which the majority of emission points are located far

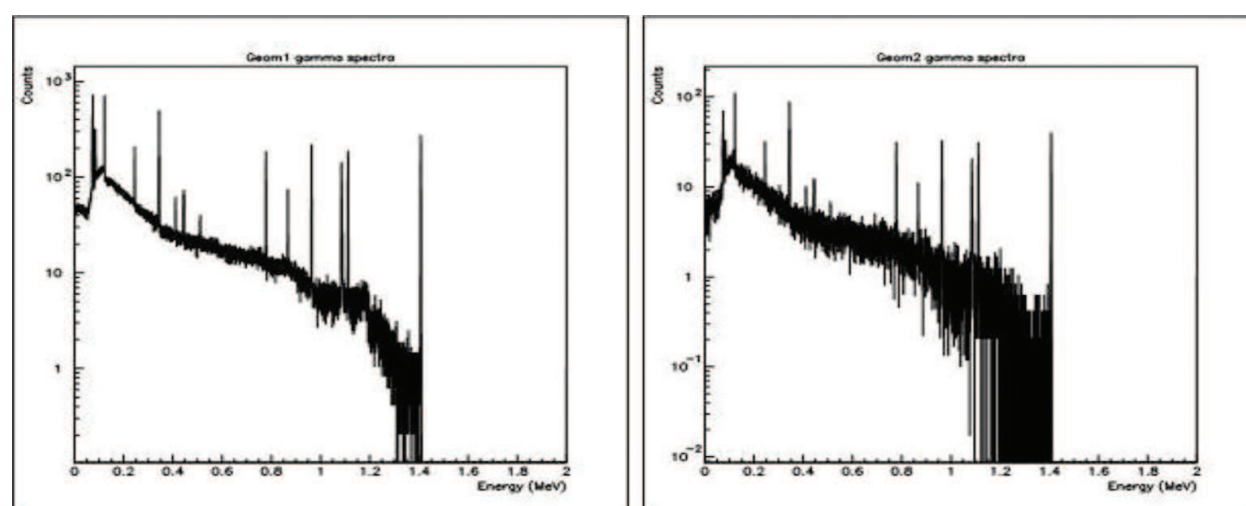
away from the detector, the fraction from the number of emitted photons that contributes to the detector signal is very small. Consequently, the number of photons that should be followed until a statistically significant number of signals will be reached should be very large, resulting in a long computation time.

In the experiment presented in this section,  $4.32\text{E}+10$  photons were simulated. All the details of the source, measurement geometry and detector were implemented in the GEANT 3.21 code. To explain the simulation process, **Figure 9** was created.



**Figure 9.** Simulation process.

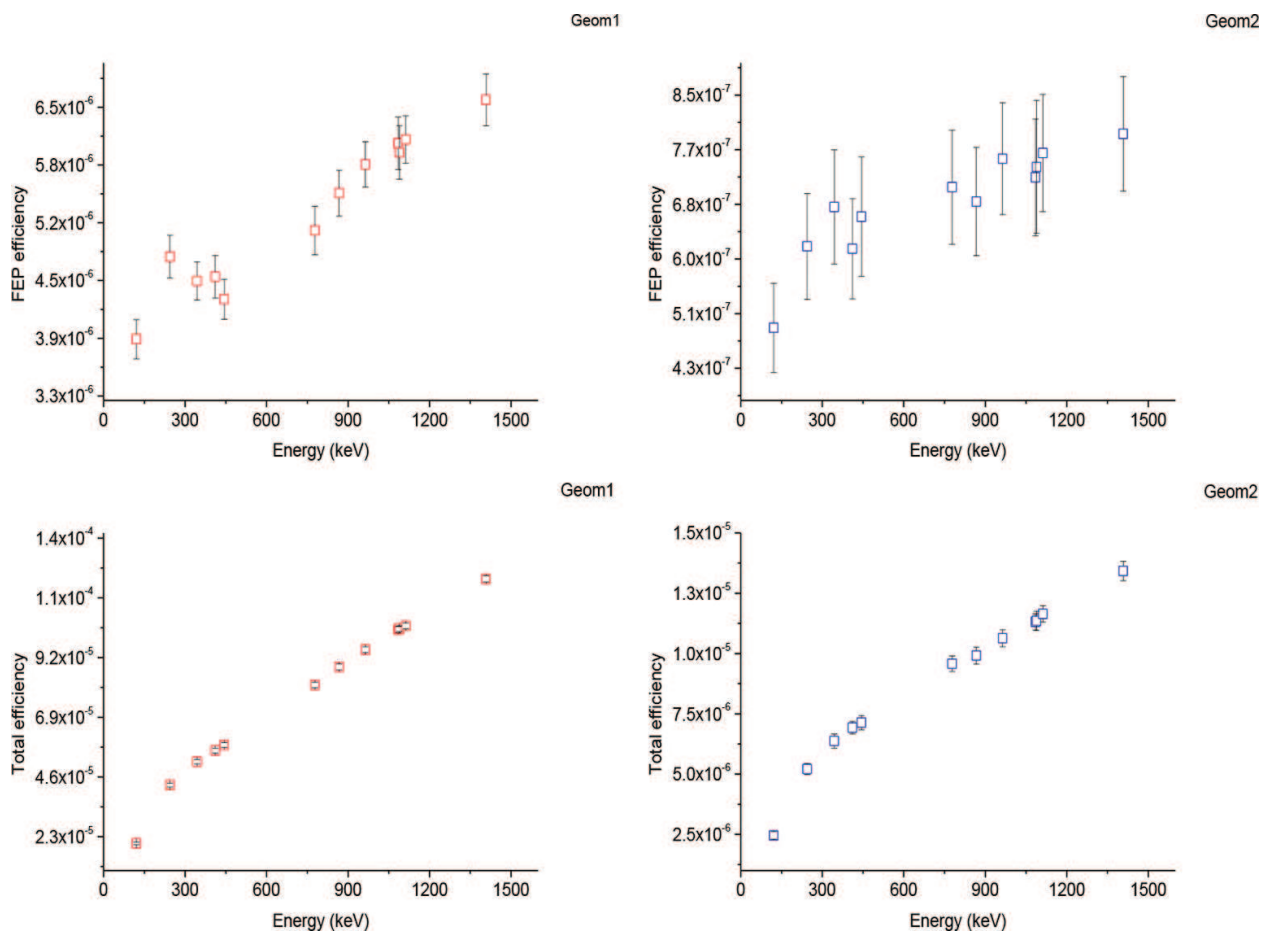
The simulations were done for the main gamma-ray photons (12 energies) emitted by  $^{152}\text{Eu}$ . In the case of Geom1,  $1.5\text{E}+09$  photons were followed for each energy, totalizing  $1.8\text{E}+10$  photons for all energies. In the case of Geom2,  $2.1\text{E}+09$  photons were simulated for each energy, representing  $2.52\text{E}+10$  photons in total for all energies. Individual spectra were recorded in separate files, and in the end, all spectra were combined with weights according to the emission probability of each gamma-ray [33]. The resulting spectra are presented in **Figure 10**.



**Figure 10.** The final spectrum for Geom1 and Geom2 geometries.

### 3.3. The detection efficiency evaluation

The detector intrinsic efficiency commonly is conditioned mainly by the material of the detector, the radiation energy and the physical thickness of the detector in the direction of the incident radiation [34]. A small dependence on source-detector distance is present due to the average path length of the radiation through the detector will amend somewhat with this area. The counting efficiencies can be classified by the nature of the events recorded. If all phenomena from the detector will be recorded, then the total efficiency will be of interest. Therefore, all interactions, no matter if the energies are small, are considered to be recorded. The peak efficiency presumes that only those interactions that deposit the full energy of the incident radiation are recorded. If the total area under the peak is integrated, then the number of full energy events can be achieved. In **Figure 11**, it represents the FEP and total peak efficiencies obtained from the simulated spectra, for Geom1 and Geom2 geometries. The fact that the efficiency in Geom2 is smaller than in Geom1 even if the second detector has a higher intrinsic efficiency is due to the smaller collimator acceptance in the case of Geom2.



**Figure 11.** The peak efficiencies and total efficiencies simulated with GEANT 3.21 for Geom1 and Geom2.

## 4. Summary and conclusions

This chapter explores the specific gamma-ray spectrometry phenomena in their deepness in different work conditions. Thus, the studies, simulations and experimental results were carried out and were presented in an integrated view in the sections of the chapter.

The utility of numerical method as it is ETNA software to compute the efficiency transfer factors for divers measurement geometries used in routine laboratory measurements was tested for NaI(Tl) and HPGe detector. Starting from a reference efficiency measured for a point source, peak efficiencies were evaluated for point sources placed at several detector-source distances, moreover for disc sources or for volume sources with different compositions and densities. This was done specifically for each detector involved in the study. The methodology shows that the efficiency transfer factors are accurately computed using ETNA software. The obtained results are valuable and can be used without restriction if all the details of the detectors and measurements are accurately known.

An important contribution in the development of gamma-ray spectrometry methods was made with the examination realized for the response function characterization of the two gamma-ray spectrometry systems: ISOCART from Ortec and WS1100 Segmented Gamma Scanner from Canberra. These systems are used especially for the measurement and characterization of radioactive waste. Based on GEANT 3.21 toolkit, a simulation program was developed to simulate the spectra expected to be obtained by the two systems, for volume sources, and for the 50–2000 keV energy range. Many spectra (hundreds) were simulated and then combined to obtain the spectrum expected in real measurements.

Considering the national regulations, the radioactivity and the nuclide composition of the waste must be identified prior to their transfer outside the site of their burial or placement in storage areas. The most important step on the characterization process is the establishment of the radionuclide content, most often achieved through non-destructive measurements (NDA). The radiological characterization of radioactive waste should ensure their correct classification and a reasonable use of interim storage and final disposal. The radioactivity overestimation leads to a reduction in waste storage capacity, and the underestimation creates problems in terms of safety. The release from regulatory control will reduce the volume of storage waste and enable their beneficial use.

Future studies are required to develop calibration techniques and evaluation of measurement for parallelepiped shape containers with radioactive waste using gamma spectrometric measurement systems. The counting geometry of parallelepiped container is completely different from the counting geometry of a small cylindrical radioactive source, and consequently, the efficiency calibration is more difficult to be estimated. Knowing the efficiency calibration, which varies greatly with the source-detector distance, the geometry and the absorption factors, is essential for the assay of radioactive waste. Because it is almost impossible to estimate the efficiency calibration curve based on the experimental measurement, simulation programs based on the Monte Carlo codes need to be developed. The evaluation of response function of HPGe detectors for parallelepiped counting geometry needs to be done.

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## References

- [1] IAEA. Methodology and technology of decommissioning. 1986; TRS No. 267.
- [2] Sima O, Lepy MC. Application of GUM Supplement 1 to uncertainty of Monte Carlo computed efficiency in gamma-ray spectrometry. *Appl. Radiation. Isot.* 2016; 109:493–499.
- [3] Debertin K, Helmer RG. Gamma and X-ray spectrometry with semiconductor detectors. North-Holland, Amsterdam. 1988.
- [4] Sima O, Arnold D. Accurate computation of coincidence summing corrections in low level gamma-ray spectrometry. *Appl. Rad. Isot.* 2000; 53:51–56.
- [5] Sima O, Arnold D. Transfer of the efficiency calibration of germanium gamma-ray detectors using the GESPECOR software. *Appl. Rad. Isot.* 2002; 56:71–75.
- [6] Sima O, Arnold D, Dovlete C. GESPECOR-A versatile tool in gamma-ray spectrometry. *J. Radioanal. Nucl. Chem.* 2001; 248:359–364.
- [7] Bruggeman M, Verheyen L, Vidmar T, Liu B. Assessing sample attenuation parameters for use in low-energy efficiency transfer in gamma-ray spectrometry. *Appl. Radiation. Isot.* 2016; 109:547–550.



- [8] Moens L, De Donder J, Lin Xi-lei, De Corte F, De Wispelaere A, Simonits A, Hoste J. Calculation of the absolute peak efficiency of gamma-ray detectors for different counting geometries. *Nucl. Instr. Meth. Phys. Res.* 1981; 187:451–472.
- [9] Lépy MC, Altzitzoglou T, Arnold D, Bronson F, Capote Noye R, Décombaz M, De Corte F, Edelmaier R, Herrera Peraza E, Klemola S, Korun M, Kralik M, Neder H, Plagnard J, Pommé S, de Sanoit J, Sima O, Ugletveit F, Van Velzen L, Vidmar T. Intercomparison of efficiency transfer software for gamma-ray spectrometry. *Appl. Radiat. Isot.* 2001; 55:493–503.
- [10] Radu D, Stanga D, Sima O. ETNA software used for efficiency transfer from a point source to other geometries. *Appl. Radiat. Isot.* 2009; 67:1686–1690.
- [11] Radu D, Stanga D, Sima O. Transfer of detector efficiency calibration from a point source to other geometries using ETNA software. *Rom. Rep. Phys.* 2010; 62:57–64.
- [12] ISO. Guide to expression of uncertainty in measurements. Geneva, Switzerland. 1993 (corrected and reprinted 1995).
- [13] Sima O, Dovlete C. Matrix effects in the activity measurement of environmental samples—implementation of specific corrections in a gamma-ray spectrometry analysis program. *Appl. Radiat. Isot.* 1997; 48:59–69.
- [14] Piton F, Lépy MC, Bé MM, Plagnard J. Efficiency transfer and coincidence summing corrections for gamma-ray spectrometry. *Appl. Radiat. Isot.* 2000; 52:791–795.
- [15] Lépy MC, Bé MM, Piton F. ETNA (Efficiency Transfer for Nuclide activity measurement): software for efficiency transfer and coincidence summing corrections in gamma-ray spectrometry. Note Technique LNHB 01/09/A. 2004.
- [16] Vidmar T, Aubineau-Laniece I, Anagnostakis MJ, Arnold D, Brettner-Messler R, Budjas D, Capogni M, Dias MS, De Geer LE, Fazio A, Gasparro J, Hult M, Hurtado S, Jurado Vargas M, Laubenstein M, Lee KB, Lee YN, Lepy MC, Maringer FJ, Medina Peyres V, Mille M, Morales M, Nour S, Plenteda R, Rubio Montero MP, Sima O, Tomei C, Vidmar G. An intercomparison of Monte Carlo codes used in gamma-ray spectrometry. *Appl. Radiat. Isot.* 2008; 66:764–769.
- [17] Choi Y, Lee KB, Kim KJ, Han J, Yi ES. Development of an optimized Compton suppression gamma-ray spectrometry system using Monte Carlo simulation. *Appl. Radiat. Isot.* (Forthcoming). doi:10.1016/j.aprasido.2015.12.058.
- [18] Jacoboni C, Lugli P, editors. The Monte Carlo method for semiconductor device simulation. Computational Microelectronics ed. Springer-Verlag Wien New York. 1989. ISSN: 0179-0307.
- [19] Geant group, editor. Physics Reference Manual, Version: Geant4 10.2 (4 December 2015). 2015. <http://geant4.web.cern.ch/geant4>.



- [20] Brun R, Bruyant F, Maire M, McPherson AC, Zanarini P. GEANT3. Geneva: CERN Data Handling Division. 1987; DDD/EE/84-1.
- [21] Agostinelli S, Allison J, Amako K, Apostolakis J, Araujo H, Arce P, Asai M, Axen D, Banerjee S, Barrand G, Behner FL, Bellagamba J, Boudreau L, Broglia Brunengo A, Burkhardt H, Chauvie S, Chuma J, Chytrcek R, Cooperman G, Cosmo G, Degtyarenko P, Dell'Acqua A, Depaola G, Dietrich D, Enami R, Feliciello A, Ferguson C, Fesefeldt H, Folger G, Foppiano F, Forti A, Garelli S, Giani S, Giannitrapani R, Gibin D, Gómez Cadenas JJ, Gonzálezq G, Gracia Abril G, Greeniaus G, Greiner W, Grichine V, Grossheim A, Guatelli S, Gumplinger P, Hamatsu R, Hashimoto K, Hasui H, Heikkinen A, Howard A, Ivanchenko V, Johnson A, Jones FW, Kallenbach J, Kanaya N, Kawabata M, Kawabata Y, Kawaguti M, Kelner S, Kent P, Kimura A, Kodama T, Kokoulin R, Kossov M, Kurashige H, Lamanna E, Lampén T, Lara V, Lefebure V, Lei F, Liendl M, Lockman W, Longo F, Magni S, Maire M, Medernach E, Minamimoto K, Mora de Freitas P, Morita Y, Murakami K, Nagamatu M, Nartallo R, Nieminen P, Nishimura T, Ohtsubo K, Okamura M, O'Neale S, Oohata Y, Paech K, Perl J, Pfeiffer A, Pia MG, Ranjard F, Rybin A, Sadilov S, Di Salvo E, Santin G, Sasaki T, Savvas N, Sawada Y, Scherer S, Sei S, Sirotenko V, Smith D, Starkov N, Stoecker H, Sulkimo J, Takahata M, Tanaka S, Tcherniaev E, Safai Tehrani E, Tropeano M, Truscott P, Uno H, Urban L, Urban P, Verderi M, Walkden A, Wander W, Weber H, Wellisch JP, Wenaus T, Williams DC, Wright D, Yamada T, Yoshida H, Zschesche D. GEANT4-a simulation toolkit. Nucl. Instr. Meth. Phys. Res. A. 2003; 506:250–303.
- [22] Briestmeister JF. MCNP—A general Monte Carlo N-particle transport code. Los Alamos National Laboratory, Report LA-13709-M. 2000.
- [23] Fassò A, Ferrari A, Roesler S, Sala PR, Ballarini F, Ottolenghi A, Battistoni G, Cerutti F, Gadioli E, Garzelli MV, Empl A, Ranft J. The physics models of FLUKA: status and recent development. eConf C0303241, MOMT005. 2003.
- [24] Berger MJ, Seltzer SM. EFTRAN-Monte Carlo code system for electron and photon transport through extended media. RISC computer code package CCC-107. Oak Ridge National Laboratory, Oak Ridge, TN. 1973.
- [25] Nelson WR, Hirayama H, Rogers DWO. The EGS4 Code System, Report SLAC-265. Stanford Linear Accelerator Center, Stanford, CA. 1985.
- [26] Hirayama H, Namito Y, Bielajew AF, Wilderman SJ, Nelson WR. The EGS5 code system, Report SLAC-R-730. Stanford Linear Accelerator Center, Stanford, CA. 2006.
- [27] Kawrakow I, Rogers DWO. The EGSnrc Code System: Monte Carlo Simulation of Electron and Photon Transport. NRCC Report PIRS-701. 2006.
- [28] Baro J, Sempau J, Fernandez-Varea JM, Salvat F. PENELOPE, an algorithm for Monte Carlo simulation of the penetration and energy loss of electrons and positrons in matter. Nucl. Instr. Meth. Phys. Res. B. 1995; 100:31–46.

- [29] Dinescu L, Vata I, Cazan IL, Macrin R, Caragheorgheopol G, Rotarescu G. On the efficiency calibration of a drum waste assay system. Nucl. Instr. Meth. Phys. Res. A. 2002; 487:661–666.
- [30] Haralambie M, Dinescu L, Sima O. New data concerning the efficiency calibration of a drum waste assay system. Part I: Experimental calibration. Rom. Rep. Phys. 2004; 56:711–720.
- [31] Toma M, Sima O, Olteanu C. Experimental and simulated studies for the calibration of a radioactive waste assay system. Nucl. Instr. Meth. Phys. Res. A. 2007; 580:391–395.
- [32] Sima O, Cazan IL, Dinescu L, Arnold D. Efficiency calibration of high volume samples using the GESPECOR software. Appl. Radiation. Isot. 2004; 61(2–3):123–127.
- [33] NUCLÉDE-LARA (web applet). Gamma and alpha spectrometry on-line library [Internet]. 2008. Available from: <http://laraweb.free.fr/>.
- [34] Knoll GF. Radiation Detection and Measurement. 4th Edition. John Wiley & Sons. 2010.

